

PATENT ABSTRACTS OF JAPAN

(11)Publication number : 05-270801

(43)Date of publication of application : 19.10.1993

(51)Int.Cl.

C01B 3/00

C01B 31/02

C22C 5/00

C22C 19/00

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(54) HYDROGEN OCCLUSION BODY

(57)Abstract:

PURPOSE: To obtain the hydrogen occlusion body by mixing fluorenes and a catalyst for hydrogenation at specific ratios.

CONSTITUTION: Known fluorenes having various kinds of carbon atoms are usable in combination of ≥ 1 kinds as the fluorenes. The more specific examples thereof includes the fluorenes of 60, 70, 76, 78, 80, 82, 84, etc., of(n)in general formula C_n . While the fluorenes having high purity are more preferable, crude products are usable as well. Generally, the use of the products having $\geq 80\%$ content of the fluorenes suffices. Various kinds of catalysts for hydrogenation are usable as the catalyst for hydrogenation and are, for example, metallic catalysts of metal colloid, etc., such as Pt colloid. The mixing ratios are preferably 1 to 200 pts.wt. catalyst for hydrogenation per 100 pts.wt. fluorenes. The fluorenes and the catalyst may be mechanically mixed and may be contact mixed by dispersing the particles of the catalyst for hydrogenation into a suitable solvent (e.g.: benzene) and dissolving or dispersing therein. The absorption of hydrogen is 50 to 250°C and the operating hydrogen pressure is usually specified to

≥ 100 Torr.

[Claim(s)]

[Claim 1] A hydrogen absorption object consisting of fullerene and catalysts for hydrogenation.

[Detailed Description of the Invention]

[0001]

[Industrial Application] If this invention is said in more detail about a hydrogen absorption object, it will carry out occlusion of a lot of [that it is lightweight and] hydrogen. It is related with the new hydrogen absorption object which can be used effective in various kinds of uses, such as absorption of hydrogen, concentration, refining, storage, conveyance, use (use as hydrogen absorption compounds, such as hydrogen gas recovery by desorption, a hydrogen gas supply agent, or a hydrogenation agent, etc.) of absorption hydrogen at any time, and application to a hydrogen cell.

[0002]

[Description of the Prior Art] In recent years, hydrogen gas is absorbed reversibly and what is called a hydrogen absorption object attracts attention as a substance which can store hydrogen compactly. The hydrogen absorption object is expected to absorb hydrogen reversibly, to be able to use, to be lightweight enough compared with the present high pressure cylinder, to be compact, and to be able to perform [various uses, such as use to reproduction, a reaction etc. by desorption of the absorbent of hydrogen, and absorption hydrogen,] stable storage and conveyance moreover especially.

[0003] However, as for all, the hydrogen absorption object known by the present is called what is called hydrogen storing metal alloy of an alloy system, for example like a LaNi alloy.

Since it is heavy since it is an alloy, and there is a problem also in stability, in particular what is satisfactory in respect of storage of hydrogen, conveyance, etc. is not obtained, but has not come to substitute for a high pressure cylinder.

Then, such a problem was fully solved, and even when it was lightweight, development of the hydrogen absorption object which consists of new construction material which can carry out occlusion of a lot of hydrogen stably was desired strongly.

[0004] By the way, the carbon cluster which has the closed shell structure of the carbon numbers 60 and 70 and 84 grades is compounded from graphite etc. as a new substance which has globular molecule structure, and the character is studied these days [for example; references, such as NATURE and Vol.347,354 (1990),]. The carbon cluster which has this special structure is also called fullerene, and is called fullerene C_{60} , the

C₇₀, the C₈₄, etc. by the carbon number which constitutes that molecular skeleton. Such fullerene is new carbon materials.

Since it is expected that unique physical properties will be shown also from having special molecular structure, research on the character and application development is advanced briskly [various kinds of fields].

[0005]Art for obtaining various kinds of derivatives from fullerene in such research and obtained research on the character and use of a derivative are also done. For example, since fullerene is unsaturation molecules, it is known that it can derive to a certain kind of hydrogenation fullerene (C₆₀H₃₆), methylation fullerene, etc. However, the actual condition is that most researches on the character or use are not made while many of these derivatives have a problem which many should solve in the synthetic art itself although it is shown that it can compound, and they had been compounded.

[0006]Then, this invention persons got the idea which is going to use the fullerene which are this new material as a hydrogen absorption object. Because, so that it may see also on the fact that C₆₀ is hydrogenated by C₆₀H₃₆, since fullerene is carbon-content children with a high degree of unsaturation, It is because it thought that it became the outstanding hydrogen absorption object which can carry out occlusion of a lot of [that it is lightweight and] hydrogen if the capability to combine a lot of hydrogen per molecule (absorption) could also be considered suddenly and this capability could be utilized effectively reversibly.

[0007]However, in order to apply fullerene as a hydrogen absorption object, ** As an improvement of the absorption process of hydrogen, as an improvement of the desorption process of the device (namely, development of the technique for making fullerene into hydrogenation fullerene efficiently) to which the rate of absorption and saturation absorbed amount of hydrogen are made to fully increase, and ** hydrogen, It is necessary to devise the device (namely, development of the technique for returning hydrogenation fullerene efficiently to the fullerene of hydrogen gas) for collecting the absorbed hydrogen efficiently as hydrogen gas, etc. Here, it may devise for using efficiently the hydrogen of which ** absorption was done for the reaction of the kind which has not necessarily been returned to hydrogen gas, either as a remedy of the desorption process of hydrogen of above **.

[0008]Thus, if pertinent arts, such as hydrogenation art of the conventional fullerene, are looked at paying attention to using fullerene as a hydrogen absorption object, no conventional technologies are those with which a case is also satisfied of the above-mentioned **, **, and ** as follows, Therefore, fullerene cannot be called art used

as a hydrogen absorption object. Even if this does not have the idea itself of utilizing fullerene effectively as a hydrogen absorption object conventionally or has an idea, it will be because the device for it was not fully made.

[0009][by which the method of obtaining $C_{60}H_{36}$ as hydrogenation art of the conventional fullerene by processing C_{60} with metal lithium and tert-butyl alcohol in liquid ammonia is known -- J. -- Phys.Chem. 94-8634-8634(1990)]. However, it is not the thing depended on hydrogen gas in this case but special reduction hydrogenation, and, needless to say, is not the art as a hydrogen absorption object from the point of absorption of hydrogen gas of the above-mentioned **, and is not significant art industrially as production technology of hydrogenation fullerene. When $C_{60}H_{36}$ compounded by this method makes it react to a special oxidizer called DDQ (2,3-dichloro-5,6-dicyanobenzoquinone), dehydration is carried out and returning to C_{60} thoroughly is known. however, by this method, hydrogen cannot be taken out by dehydrogenation, therefore it carries out and possibility is not shown at all in respect of [which is not the art of the above-mentioned **] the application to a reaction with others general only by returning the strong oxidizer special also as art of the above-mentioned **. That is, in this case, even if it notes only the point of **, it is clear to need the new device for fully improving the reactivity of hydrogenation fullerene and using the absorbed hydrogen effectively more efficiently.

[0010]On the other hand, contacting C_{60} to a hydrogenation catalyst is known as organic palladium polymer $C_{60}Pd_n$ [the 2nd C_{60} synthesis symposium lecture proceedings p20 (1992)]. However, in this report, having a hydrogenation function to the compound which only has a double bond is reported, and this compound contacted to fullerene (C_{60}) and a hydrogenation catalyst (Pd) is a request.

[0011]As mentioned above, the art of applying fullerene to a hydrogen absorption object including C_{60} was not known conventionally.

[0012]

[Problem(s) to be Solved by the Invention]This invention is made in view of said situation.

[0013]The purpose of this invention can carry out occlusion of a lot of [that it is lightweight and] hydrogen effectively, is advantageous to absorption of hydrogen, and storage and conveyance, and there is in moreover providing the new hydrogen absorption object remarkable and practically, useful effective in a hydrogenation reaction agent, a hydrogen supply agent, etc. in the state of recovery use or hydrogen absorption by desorption of the absorbed hydrogen.

[0014]

[Means for Solving the Problem]The fullerene of this invention persons is [that said purpose should be attained] lightweight compared with the conventional hydrogen storing metal alloy from consisting of carbon, And paying attention to having the capability to combine a lot of hydrogen per molecule (absorption), research was repeated wholeheartedly that it should improve [fullerene / including C₆₀ / various kinds of] about the aforementioned **, **, and/or **. As a result, a substance or a substance system of various kinds of fullerene and various kinds which combines a hydrogenation catalyst satisfied the aforementioned **, **, and **, and found out becoming the outstanding hydrogen absorption object which can fully attain said purpose. This invention persons came to complete this invention based on these knowledge.

[0015]That is, a hydrogen absorption object, wherein this invention consists of fullerene and catalysts for hydrogenation is provided.

[0016]In this invention, it is one-sort independent or the fullerene which have various kinds of carbon numbers, such as publicly known fullerene, as said fullerene can be used combining two or more sorts. As an example of these fullerene, it is expressed with general formula C_n and n in this type can mention fullerene of various kinds of carbon numbers, such as 60, 70, 76, 78, 80, 82, and 84, for example. Of course, these should show a case in the state where occlusion of the hydrogen has not been carried out, and a part or all of these fullerene should be noticed about a point of changing to hydrogenation fullerene, in the state where a hydrogen absorption object of this invention is absorbing hydrogen (occlusion). That is, a hydrogen absorption object of this invention may be made to form using various kinds of fullerene (hydrogenation fullerene etc.) which combined or contained hydrogen to saturation selectively. With a hydrogen absorption object of this invention, since absorption, desorption, or consumption (use) of hydrogen is performed reversibly, this, Even if it uses hydrogen content fullerene, such as hydrogenation fullerene, it is because it becomes the hydrogen absorption object outstanding like a case where it prepares using fullerene as a result by using the hydrogen for desorption or a reaction.

[0017]Although what has high purity is preferred as for said fullerene, a thing of a high grade may not necessarily be used for it, either, and it may use what contains other ingredients (for example, soot-like carbon etc.) in the range which does not check the purpose of this invention. For example, the crude fullerene containing various kinds of fullerene (for example, C₆₀, C₇₀, etc.) produced from coal-for-coke-making matter, such as graphite, by carrying out solvent extraction of the fullerene content soot-like substance compounded by arc discharge, laser radiation reaction, etc. can be used conveniently. Of course, the fullerene of a high grade which isolated by the refined

refining fullerene, chromatography separation, etc. can also be used. The amount of absorption (occlusion) of hydrogen per unit weight can be enlarged, so that fullerene concentration in a fullerene inclusion to be used is high. On the other hand, since refining cost of fullerene becomes large, a price of a hydrogen absorption object of the part this invention also becomes high, so that purity of fullerene is made high. Generally, if content of fullerene uses not less than 80% of thing, it is enough, and ** which selects purity of the fullerene suitably used according to cost or the purpose of use in this range is preferred.

[0018]It is usable in various kinds of catalysts for hydrogenation, such as what restriction in particular does not have, and is used or proposed as a hydrogenation catalyst of publicly known hydrocarbon etc. as said hydrogenation catalyst used in order to make a hydrogen absorption object of this invention form. As such a catalyst for hydrogenation, for example Cr, Fe, Co, nickel, If there is a catalyst of various gestalten which consist of various kinds of metal including transition metals, such as Mo, Ru, Rh, Pd, W, Re, Os, Ir, and Pt, and a typical thing of them is illustrated, For example, metal system catalysts, such as metallic colloid represented with Pt colloid, a Raney nickel catalyst, the Raney ruthenium, Raney cobalt, etc., Platinum black, palladium black, ruthenium black, rhodium black, A metallic-oxide system catalyst represented with rhenium black, chrome oxide, molybdenum oxide, etc., To metallic-compounds system catalysts, such as a metallic sulfide system catalyst represented, various kinds of metal complex system catalysts, etc., such as a molybdenum sulfide and rhenium sulfide, and a pan. these metal or metallic compounds -- various kinds of carriers (for example, a carbon system carrier, such as activated carbon,) Various kinds of supported type catalysts supported to oxide stock carriers, such as silica, alumina, a silica alumina, clay, diatomaceous earth, various composition, or natural silicate, etc. (for example, various things, such as support Pd / carbon, Ru/carbon, nickel/diatomaceous earth, Pd/silica, can be mentioned.) Pd/carbon, Ru/carbon, nickel/diatomaceous earth, etc. can be mentioned as what can be especially used conveniently also in these, for example. These hydrogenation catalysts may be used by an one-sort independent, and two or more sorts can be mixed, or can be composite-ized, and they can also use them together.

[0019]A hydrogen absorption object of this invention is formed by mixing at least, so that said hydrogenation catalyst may contact said fullerene.

[0020]Since the suitable mixing ratio of said fullerene and said hydrogenation catalyst changes with kinds and mixed form voice of a hydrogenation catalyst to be used, it cannot be defined uniformly, but it is preferred for it to select in proportion that a hydrogenation catalyst usually serves as one to 200 weight section to fullerene 100

weight section to be used. There may be so few the rates that the hydrogenation activity of a hydrogenation catalyst to be used is generally high.

[0021]As contact mixed form voice of said fullerene and a hydrogenation catalyst, various forms are possible. For example, a method which carries out mechanical [of said fullerene and said hydrogenation catalyst] (it mixes physically), and is used as a mixed granule child and a composite particle of a solid state, A method which supports an active metal ingredient of said hydrogenation catalyst into a mixture with fullerene or fullerene, and carrier particles, and carries out contact mixing, Particles of said hydrogenation catalyst are distributed in this solvent using a suitable solvent, A soluble hydrogenation catalyst can be dissolved into a method which makes it dissolve and/or distribute and carries out contact mixing of said fullerene, and a solvent, and various methods, such as a method which make it dissolve and/or distribute and carries out contact mixing of said fullerene, can be adopted. Thus, the hydrogen absorption object of this invention can use and carry out contact mixing of said fullerene and said hydrogenation catalyst through a solvent, can also make it form, and. Although contact mixing may be carried out without using a solvent and it may be made to form, since a method to which this is contacted as a medium using a suitable solvent has good contacting efficiency, generally it is preferred.

[0022]As said solvent, for example Aromatic solvents, such as benzene, toluene, xylene, and mesitylene, Although alkanes, such as alicyclic hydrocarbon solvents, such as cyclohexane, a methylcyclohexane, dimethylcyclohexane, and trimethylcyclohexane, hexane, heptane, and octane, can be mentioned, it is not necessarily limited to these. These solvents may be used as an one-sort independent solvent, and may be used as a mixed solvent which consists of two or more sorts.

[0023]In the range which does not check the purpose of this invention if needed, other ingredients other than the above are made to contain, and it may be sufficient, and may combine or may be used for a hydrogen absorption object of this invention.

[0024]A hydrogen absorption object of this invention is effectively used by repeating operation with absorption (occlusion) and absorbed dehydration of hydrogen of hydrogen.

[0025]Absorption of hydrogen can be performed by contacting hydrogen gas to this occlusion body. Although temperature and hydrogen pressure in the case of this hydrogen absorption should just determine suitably a kind of hydrogenation catalyst, existence, a kind of solvent which are included in this occlusion body in consideration of other conditions, they are performed in the range of temperature as which that catalyst functions as a hydrogenation catalyst at least, and hydrogen pressure. Absorption

(hydrogenation of fullerene) speed is so quick that hydrogen pressure is generally high, and saturation absorption hydrogen quantity also increases. Generally a rate of absorption becomes quick so that temperature is high, but since a balance is based on the dehydration side under the same hydrogen pressure, if temperature is made not much high, in order to obtain sufficient saturation absorption hydrogen quantity, hydrogen pressure becomes high so much. When using a solvent, it is preferred to define conditions, such as temperature, in consideration of steam pressure of a solvent. Thus, since temperature and hydrogen pressure of hydrogen absorption suitable in the case change with other various conditions besides a kind of hydrogenation catalyst and temperature is mutually related to hydrogen pressure, cannot define each uniformly, but. It is preferred to select operating temperature suitably to temperature of the range of 50-250 **, and to usually select preferably 100 or more Torr of operation hydrogen pressure suitably generally to ** of the range of $2\text{kg/cm}^2 - 150\text{ kg/cm}^2$.

[0026]On the other hand, various forms can perform according to the purpose, desorption, i.e., dehydration, of hydrogen from an occlusion body which absorbed hydrogen (occlusion). That is, a method which collects hydrogen gas according to dehydration, and an occlusion body which absorbed hydrogen (occlusion) can be used for a hydrogen donor, a hydrogenation reaction agent, etc. and a direct reaction, and various kinds of dehydration methods, such as a method which carries out dehydration, can be applied by making hydrogen of this absorber react to other reactants, or making it move. In other words, a hydrogen absorption object of this invention uses absorption (occlusion) and desorption for the hydrogen, A thing in the state where could use in order to obtain hydrogen gas of a high grade at any time, and hydrogen was absorbed (occlusion) can also be used for a hydrogenation agent etc. and other useful synthetic reactions, and can be further used also for a use of a hydrogen carrier etc. Therefore, since conditions in the case of this dehydration change remarkably with uses, it cannot set uniformly, but this dehydration is carried out on conditions on which a hydrogenation catalyst contained in this occlusion body at least has a function as a dehydrogenation catalyst. With the dehydration function of this catalyst, desorption as hydrogen gas can be promoted and other hydrogenation reactions and hydrogen shift processes over a reactant can be promoted.

[0027]In collecting absorption (occlusion) hydrogen as hydrogen gas, It can attain like [in the case of the usual hydrogen storing metal alloy] by making operating temperature of this dehydration higher than temperature at the time of hydrogen absorption (occlusion), making operating pressure (hydrogen content pressure) of dehydration lower than hydrogen pressure at the time of hydrogen absorption

(occlusion), or performing them simultaneously further. Hydrogen gas according to this dehydration a suitable temperature and ** (hydrogen content pressure) for a recovery sake, Since it is based on conditions, i.e., an occlusion rate of hydrogen, etc., at the time of a kind of hydrogenation catalyst, and absorption (occlusion) of hydrogen (degree of hydrogenation of fullerene) and ** is mutually related to temperature, cannot set uniformly, but. Generally, operating temperature is suitably selected to temperature of the range of 60-280 **, and it is operating pressure (hydrogen content pressure). Selecting suitably is [below 50 kg/cm²] usually suitable for ** of the range of 0kg/cm² - 20kg/cm² preferably. A desorption rate as hydrogen gas becomes large, so that ** (hydrogen content pressure) is so low that this temperature is high.

[0028]namely, -- making a cycle of absorption (occlusion) of hydrogen gas, and desorption recovery of hydrogen gas be the same as that of a case of the conventional hydrogen storing metal alloy in a hydrogen absorption object of this invention -- normal operation methods, such as the temperature swinging method, the pressure swinging method, and the thermal stress simultaneous swinging method, -- suitable -- a line -- things are made. Since an absorption (occlusion)-desorption cycle of hydrogen gas can be effectively performed only on swing of a pressure, without changing temperature since the pressure effect in this cycle is large in the case of a hydrogen absorption object of this invention, it is remarkably advantageous in respect of energy cost and simplicity of operation.

[0029]What is necessary is on the other hand, just to define conditions, such as temperature, suitably in consideration of conditions, such as a reaction of the request, in carrying out dehydration to other purposes, such as a hydrogenation reaction, using an occlusion body which absorbed hydrogen (occlusion). Since it depends remarkably for the purpose, such as a reactant and a kind of the reaction itself, also at a low temperature, a dehydration speed high enough is obtained depending on a reaction, and conditions in this case can attain the purpose.

[0030]A hydrogen absorption object of this invention can be stably saved in the state where this was made to absorb hydrogen (occlusion), and can be carried. If storage (conveyance) of hydrogen with this hydrogen absorption object is a temperature low enough, it can be stably performed also under un-living together of hydrogen gas, but since dehydration will advance so easily if temperature becomes high, it is desirable to save in that case at a suitable well-closed container. however, since absorption (occlusion) of hydrogen is stored in a form of a comparatively stable compound called hydrogenation fullerene in the case of a hydrogen absorption object of this invention, unless temperature is made remarkably high, it does not necessarily save at high

pressure vessels-proof, such as a high pressure cylinder, -- also coming out -- there is an advantage that it can save and carry safely. For example, even if it dedicates to the usual container and does preservation and conveyance of near a room temperature, generally trouble is not produced.

[0031]A lightweight carbon system material is used for a hydrogen absorption object of this invention compared with conventional alloys called fullerene, Since the amount of hydrogenation catalyst used is good at least by a daily dose about a catalyst and a lot of hydrogen is absorbed per one molecule of fullerene like $C_{60}H_{18}$ or $C_{60}H_{36}$, for example in fullerene C_{60} (combination), Even if lightweight, a lot of hydrogen is absorbable (occlusion). Therefore, it can store and carry safely compactly and lightweight, without using a heavy container which it is remarkably advantageous to storage, conveyance, etc. as compared with the conventional hydrogen storing metal alloy, and is called a high pressure cylinder also from this point. Carry out occlusion of a lot of [that a hydrogen absorption object of this invention is lightweight, and] hydrogen, and Thus, absorption of hydrogen, concentration, refining, storage, conveyance, and use (hydrogen gas recovery by desorption, and a hydrogen gas supply agent.) at any time in various forms of absorption hydrogen or use as hydrogen absorption compounds, such as a hydrogenation agent, application to a hydrogen cell, etc. -- etc. -- it is the outstanding hydrogen absorption object which can be used in favor of various kinds of uses, and can be used conveniently for various kinds of fields.

[0032]

[Example]Hereafter, although the example of this invention explains this invention more concretely, this invention is not limited to these examples.

[0033]500 mg of crude fullerene which contains 85 % of the weight and C_{70} for example $1C_{60}$ 15% of the weight was dissolved in 500 ml of toluene, 1.0 g was added for a 5% ruthenium / carbon (50%wet) as a hydrogenation catalyst, and the hydrogen absorption object was acquired. It was made to react to this hydrogen absorption object at the temperature of 100 ** in hydrogen pressure 50 kg/cm² for 6 hours, and occlusion of hydrogen was performed. When mass-spectrum measurement of reaction mixture was performed, both fullerene C_{60} and C_{70} were hydrogenated, and the main output was $C_{60}H_{36}$. Next, the main output was C_{60} when the mass spectrum of reaction mixture after making toluene distribute residue after removing a solvent and heating at 120 ** under atmospheric pressure for 3 hours was measured. Generating of 180 ml of hydrogen was checked by this reaction.

[0034]Replaced example 2 hydrogenation catalyst with the cobalt system meltable catalyst, 100 ml of methylcyclohexanes were made to distribute the fullerene

constituent 1.0g, and the hydrogen absorption object was acquired. Occlusion of hydrogen was carried out to this hydrogen absorption object at hydrogen pressure 20kg/cm² and the temperature of 50 °C for 3 hours. The hydrogen consumption at this time was 0.47 l. When this reactant was heated to 110 °C under the atmosphere, 0.44 l. of hydrogen was collected. Next, when occlusion of hydrogen was again carried out to the hydrogen absorption object after dehydration at hydrogen pressure 20kg/cm² and the temperature of 50 °C for 3 hours, the amount of consumption (absorbed amount) of hydrogen is 0.44 l., and hydrogen had the whole quantity collected by heating under the atmospheric pressure in the above-mentioned conditions. Degradation was not seen even if it repeated the above-mentioned reaction (occlusion and dehydration).

[0035]

[Effect of the Invention]According to this invention, can carry out occlusion of a lot of [that it is lightweight and] hydrogen effectively, and are advantageous to absorption of hydrogen, and storage and conveyance, And the hydrogen absorption object remarkable and practically, useful which can be suitably used also as a hydrogenation reaction agent, a hydrogen supply agent, etc. in the state of the recovery use by desorption of the absorbed hydrogen or hydrogen absorption can be provided.

[Industrial Application]If this invention is said in more detail about a hydrogen absorption object, it will carry out occlusion of a lot of [that it is lightweight and] hydrogen, It is related with the new hydrogen absorption object which can be used effective in various kinds of uses, such as absorption of hydrogen, concentration, refining, storage, conveyance, use (use as hydrogen absorption compounds, such as hydrogen gas recovery by desorption, a hydrogen gas supply agent, or a hydrogenation agent, etc.) of absorption hydrogen at any time, and application to a hydrogen cell.

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2. **** shows the word which can not be translated.

3. In the drawings, any words are not translated.